INSECTICIDE RESIDUES IN TOTAL DIET SAMPLES

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ABSTRACT

Pesticide residues were detected in total diet samples randomly collected from Cairo. More than 23 pesticide residues and their degradation products were detected by GLC. Endrin, dieldrin, lindane and total DDT were the main residues detected in most samples. The amount of pesticide residues consumed by the average person in the average diet was calculated. Results indicated that the feeding habits and behavior of the Egyptian people play an important role in their daily intake of pesticide residues. High quantities of drinking water and bread are consumed daily and these would be the main sources of intake. In fact bread was the source of more than 50% of the pesticide daily intake.

The daily intake of endrin, dieldrin, lindane and total DDT from bread was 0.0960, 0.0624, 0.5280 and 0.5760 mg/person.

The total daily intake of pesticide residues was 0.1671, 0.0555, 0.7018 and 0.0578 mg endrin, dieldrin, lindane and total DDT/person respectively, while the acceptable daily intake for the fore-mentioned insecticides is 0.014, 0.007, 0.7 and 1.4 mg/person.

INTRODUCTION

Egypt used 617507 metric tons of pesticides in the period between 1952 and 1984 [Abdel-Gawaad, 1985]. Nearly 50% of this quantity found its way into the soil and is a permanent source of pesticides persisting and affecting plants, soil fertility, air water and useful fauna [Abdel-Gawaad, 1981 a & b].

Measurable amounts of pesticide residues in our food present a variety problems. Edwards, (1973), reported that organochlorine insecticides in use for more than four decades in agriculture and public health programmes all over the world, have caused one of the most serious environmental problems and are commonly detected in air, soil, water, aquatic and terrestrial wild life and in different food items. Szokolay et al., (1977), reported that these compounds reach the human body in the daily diet and many of them accumulate in adipose tissue. Al- Omar et al., (1986), reported that contamination of human milk with residues of organochlorine insecticides represents a major problem caused by pesticide environmental pollution. Fluctuation in the residue levels was obviously due to variations in the daily dietary intake of residues and variations in fat content of maternal milk.

The aim of this study is to assess the amount of pesticide residues consumed by the average person in the average diet. Also, to throw light on the feeding habits and behavior of the Egyptian people and the relation between those and the international guide line values for pesticide residues.
MATERIALS AND METHODS

Samples
Fifty samples of total diet were collected. Each sample comprised Egyptian bread, meat (beef, fish, poultry), vegetables (tomatoes, potatoes), milk and milk products, oil and fruits. Samples were kept at 18°C until analysis.

1- Egyptian bread, fruits, vegetables, meat and dairy products

A- Extraction
Representative samples were finely chopped and were mixed prior to subsampling. Subsample of 50g each were then macerated for 1 min. with 2 ml of propylene carbonate per gram of samples. Vacuum-filtered through glass wool and a layer of granular unhydrous sodium sulfate. The filtered extract was redried over approximately 20g of sodium sulfate.

B- Clean-up
Column chromatography was used to separate the organic chlorine and organophosphorous compounds from propylene carbonate extracts.

50g of deactivated florasil were added to a chromatographic column (25mm i.d. X 400mm) containing 5g of sodium sulfate. An additional 5g of sodium sulfate were added at the top of the florasil. The column was prewashed with 50 ml of petroluem ether, and the washings were discarded.

5ml of extract, representing 2.5 g of samples were transferred to the column and allowed to penetrate the upper portion of the florasil. Elution was then performed with 200 ml volumes of 7 and 25% diethylether in petroluem ether; one fraction contained organic chlorine compounds and the other contained organothiophosphorous compounds. Each 200 ml fraction was collected in a Kuderna-Danish flask for concentration to dryness. The concentrates were finally dissolved in 1 ml acetone for G.C. analysis.

2- Milk
A modified AOAC method reported by Zazuki et al., (1979), was used. 50 ml of milk were mixed with 0.5g potassium oxalate and 50 ml ethanol. Fat was extracted 3 times with 50 ml portions of ethyl ether: petroluem ether (1:1). The combined organic layers were washed with water, dried and concentrated to 25 ml. 1 ml concentrate was used to determine the extract weight. 10 ml was transferred to a separatory funnel and the pesticides were extracted 4 times with 20 ml portions of acetonitrile saturated with petroluem ether.

Combined acetonitrile extracts were diluted with 450 ml of water and the aqueous phase was reextracted twice with 8 ml portions of petroluem ether. Petroleum ether solutions were combined, dried and concentrated to 10 ml. A 4 ml extract was submitted to florasil column for clean-up as previously described.

3- Drinking Water
A- Extraction
500 ml sample was extracted with 25 ml of benzene in a single extraction by shaking in a separatory funnel for 2 min. The separated extract was concentrated to 1 ml by blowing a stream of air over the benzene extract in a fume cubbord.
Gas Chromatography Determinations
Analyses were performed on a gas liquid chromatograph equipped with electron capture detector.

Columns
Glass, 6 mm OD by 4 mm ID, 183 cm-long packed with OV-17 was used.

Carrier Gases
Pure nitrogen at a flow rate of 30 ml/min.
Pure hydrogen at a flow rate of 60 ml/min.
Air at the flow rate of 300 ml/min.

Temperature

RESULTS AND DISCUSSION

Data were collected from a statistical survey conducted by the Egyptian Nutrition Institute (Ministry of Public Health), about the quantity of food in the daily diet of Egyptian families, adults, toddlers and infants.

The average diet/person/day is tabulated in table 1. The Egyptians drinks a large quantity of water and consume large quantities of bread in comparison with people in developed countries. Bread is the main food (48%), while meat constitutes only 3.3% of the Egyptian diet.

Table 1: Average composition of the Egyptian person’s total daily diet.

<table>
<thead>
<tr>
<th>Food group</th>
<th>Average Weight (g/day)</th>
</tr>
</thead>
<tbody>
<tr>
<td>drinking water</td>
<td>2810 *</td>
</tr>
<tr>
<td>whole milk</td>
<td>83.3 **</td>
</tr>
<tr>
<td>milk products</td>
<td>16.7 **</td>
</tr>
<tr>
<td>meat, fish or poultry</td>
<td>33.3 **</td>
</tr>
<tr>
<td>bread (cereal grains)</td>
<td>480.0 **</td>
</tr>
<tr>
<td>potatoes</td>
<td>100.0 **</td>
</tr>
<tr>
<td>vegetables</td>
<td>116.7 **</td>
</tr>
<tr>
<td>fruits and fruit juices</td>
<td>73.3 **</td>
</tr>
<tr>
<td>oils and fats</td>
<td>13.3 **</td>
</tr>
<tr>
<td>sugar and adjuncts</td>
<td>86.7 **</td>
</tr>
</tbody>
</table>

* calculated
** data from the Egyptian Nutrition Institute.

Table 2 contains residue values found for some chlorinated hydrocarbons detected in the daily diet samples.
Out of 50 water samples, 41 samples contained insecticide residues. 9 samples were free from endrin, dieldrin, lindane and total DDT. The minimum and maximum residue levels of endrin, dieldrin, lindane and total DDT were 0.001-0.015, 0.000-0.004, 0.001-0.005 and 0.030-0.054 ppm respectively.

Table 2: Organochlorine insecticide residues in individual food groups of the total diet [ppm].

<table>
<thead>
<tr>
<th>Food Group</th>
<th>endrin</th>
<th>dieldrin</th>
<th>lindane</th>
<th>ΣDDT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drinking Water</td>
<td>min. 0.001</td>
<td>0.00</td>
<td>0.001</td>
<td>0.030</td>
</tr>
<tr>
<td></td>
<td>mean 0.01</td>
<td>0.003</td>
<td>0.004</td>
<td>0.043</td>
</tr>
<tr>
<td></td>
<td>max. 0.015</td>
<td>0.004</td>
<td>0.005</td>
<td>0.054</td>
</tr>
<tr>
<td>Whole Milk</td>
<td>min. 0.001</td>
<td>0.00</td>
<td>0.002</td>
<td>0.040</td>
</tr>
<tr>
<td></td>
<td>mean 0.028</td>
<td>0.016</td>
<td>0.022</td>
<td>0.334</td>
</tr>
<tr>
<td></td>
<td>max. 0.035</td>
<td>0.021</td>
<td>0.032</td>
<td>0.391</td>
</tr>
<tr>
<td>Dairy Products</td>
<td>min. 0.00</td>
<td>0.000</td>
<td>0.003</td>
<td>0.061</td>
</tr>
<tr>
<td></td>
<td>mean 0.03</td>
<td>0.050</td>
<td>0.061</td>
<td>0.925</td>
</tr>
<tr>
<td></td>
<td>max. 0.05</td>
<td>0.065</td>
<td>0.068</td>
<td>1.200</td>
</tr>
<tr>
<td>Meat, Fish, Poultry</td>
<td>min. 0.030</td>
<td>0.002</td>
<td>0.004</td>
<td>0.062</td>
</tr>
<tr>
<td></td>
<td>mean 0.119</td>
<td>0.011</td>
<td>0.092</td>
<td>0.616</td>
</tr>
<tr>
<td></td>
<td>max. 0.390</td>
<td>0.035</td>
<td>0.130</td>
<td>0.688</td>
</tr>
<tr>
<td>Bread (cereal grains)</td>
<td>min. 0.02</td>
<td>0.07</td>
<td>0.03</td>
<td>0.060</td>
</tr>
<tr>
<td></td>
<td>mean 0.20</td>
<td>0.130</td>
<td>1.1</td>
<td>1.20</td>
</tr>
<tr>
<td></td>
<td>max. 0.36</td>
<td>0.270</td>
<td>2.30</td>
<td>2.18</td>
</tr>
<tr>
<td>Potatoes</td>
<td>min. 0.03</td>
<td>0.08</td>
<td>0.001</td>
<td>0.03</td>
</tr>
<tr>
<td></td>
<td>mean 0.21</td>
<td>0.134</td>
<td>0.30</td>
<td>1.10</td>
</tr>
<tr>
<td></td>
<td>max. 0.63</td>
<td>0.230</td>
<td>0.65</td>
<td>2.13</td>
</tr>
<tr>
<td>Vegetables (tomatoes)</td>
<td>min. 0.00</td>
<td>0.02</td>
<td>0.03</td>
<td>0.20</td>
</tr>
<tr>
<td></td>
<td>mean 0.01</td>
<td>0.12</td>
<td>0.21</td>
<td>0.70</td>
</tr>
<tr>
<td></td>
<td>max. 0.21</td>
<td>0.30</td>
<td>0.45</td>
<td>0.29</td>
</tr>
<tr>
<td>Fruits and Fruit Juices</td>
<td>min. 0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>mean 0.003</td>
<td>0.001</td>
<td>0.002</td>
<td>0.05</td>
</tr>
<tr>
<td></td>
<td>max. 0.010</td>
<td>0.050</td>
<td>0.045</td>
<td>0.16</td>
</tr>
<tr>
<td>Oil and Fats</td>
<td>min. 0.20</td>
<td>0.08</td>
<td>0.06</td>
<td>0.80</td>
</tr>
<tr>
<td></td>
<td>mean 1.02</td>
<td>0.13</td>
<td>0.14</td>
<td>1.402</td>
</tr>
<tr>
<td></td>
<td>max. 2.50</td>
<td>1.70</td>
<td>3.08</td>
<td>3.20</td>
</tr>
<tr>
<td>Sugar and Adjuncts</td>
<td>min. 0.00</td>
<td>---</td>
<td>0.00</td>
<td>0.000</td>
</tr>
<tr>
<td></td>
<td>mean 0.002</td>
<td>---</td>
<td>0.003</td>
<td>0.001</td>
</tr>
<tr>
<td></td>
<td>max. 0.03</td>
<td>---</td>
<td>0.010</td>
<td>0.020</td>
</tr>
</tbody>
</table>
The daily intake of residues through drinking water for a person of 70 kg was 0.00281, 0.0008, 0.0112 and 0.1208 mg of endrin, dieldrin, lindane and total DDT respectively (Table 3).

Table 3: Pesticide residues daily intake [mg] for an Egyptian person.

<table>
<thead>
<tr>
<th>Food Group</th>
<th>endrin</th>
<th>dieldrin</th>
<th>lindane</th>
<th>DDT</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drinking Water</td>
<td>0.00281</td>
<td>0.0008</td>
<td>0.0112</td>
<td>0.1208</td>
</tr>
<tr>
<td>Milk</td>
<td>0.0023</td>
<td>0.0013</td>
<td>0.0018</td>
<td>0.0278</td>
</tr>
<tr>
<td>Dairy Products</td>
<td>0.0005</td>
<td>0.0008</td>
<td>0.0010</td>
<td>0.0154</td>
</tr>
<tr>
<td>Meat, Fish, Poultry</td>
<td>0.0040</td>
<td>0.0004</td>
<td>0.0031</td>
<td>0.0205</td>
</tr>
<tr>
<td>Bread (Cereal Grains)</td>
<td>0.0960</td>
<td>0.0624</td>
<td>0.5280</td>
<td>0.5760</td>
</tr>
<tr>
<td>Potatoes</td>
<td>0.0210</td>
<td>0.0154</td>
<td>0.0300</td>
<td>0.1100</td>
</tr>
<tr>
<td>Vegetables</td>
<td>0.0012</td>
<td>0.0140</td>
<td>0.0245</td>
<td>0.0816</td>
</tr>
<tr>
<td>Fruits and Fruit Juices</td>
<td>0.0002</td>
<td>0.0007</td>
<td>0.0001</td>
<td>0.0037</td>
</tr>
<tr>
<td>Oil and Fats</td>
<td>0.0136</td>
<td>0.0017</td>
<td>0.0018</td>
<td>0.0019</td>
</tr>
<tr>
<td>Sugar and Adjuncts</td>
<td>0.0002</td>
<td>-</td>
<td>0.0003</td>
<td>0.0001</td>
</tr>
<tr>
<td>Total Acceptable daily intake for a Person of 70 Kg.</td>
<td>0.1671</td>
<td>0.0955</td>
<td>0.7018</td>
<td>0.9578</td>
</tr>
</tbody>
</table>

The daily intake of the fore-mentioned insecticide residues with the total diet was 0.1390, 0.0947, 0.6906 and 0.8370 respectively.

More than 50% of the daily intake of insecticide residues absorbed by the Egyptian person comes from bread. The habit and behavior of the Egyptian people shows that they consume daily nearly half a kg of cereal grains (as bread) which can be considered as the main source of pesticide residues.

The daily intake of endrin, dieldrin, lindane and total DDT through bread is 0.0960, 0.0624, 0.5280 and 0.5760 mg/person. All the other food groups are responsible for daily intakes of only 0.0430, 0.0323, 0.1826 and 0.2610 mg of endrin, dieldrin, lindane and total DDT respectively.

It is clear from the data in table 3 that the main source of insecticide residues for Egyptian people is cereal grains (as bread) followed by drinking water, potatoes, vegetables and oil respectively.

Milk, milk products and all types of meat come as a third category. The total daily intake of pesticide residues was 0.1617, 0.0955, 0.7018 and 0.9578 mg of endrin, dieldrin, lindane and total DDT respectively/person.

These levels are acceptable only in the case of lindane and total DDT while the levels were very high in the case of endrin and dieldrin (Table 4).

Beside the fore-mentioned pesticide residues, traces of aldrin, chlordane, HCH, were detected as well as 23 unknown compounds. Putting into consideration that there is an additional source of the present pesticides load in the human body, resulting from air, dust and tobacco. These results indicate that pesticide residues in Egyptian food may be the source of a variety of serious health problems. Certain publications revealed the high number of deaths caused by cancer in Egypt [Esmatt, 1985]. For those reasons Egyptian maximum residue limits for each food group should be established individually. Based as closely as practical to WHO/FAO recommendations in the context of prevailing environmental, social, economic and cultural local conditions.
Table 4: List of Codex maximum residue limits (mg/kg)

<table>
<thead>
<tr>
<th>Food Group</th>
<th>endrin</th>
<th>dieldrin</th>
<th>lindane</th>
<th>2007</th>
</tr>
</thead>
<tbody>
<tr>
<td>Drinking Water</td>
<td>-</td>
<td>0.0003</td>
<td>0.003</td>
<td>0.001*</td>
</tr>
<tr>
<td>Milk</td>
<td>0.0008</td>
<td>0.006</td>
<td>0.01</td>
<td>0.5</td>
</tr>
<tr>
<td>Dairy Products</td>
<td>0.1</td>
<td>-</td>
<td>2</td>
<td>5</td>
</tr>
<tr>
<td>Fish</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>5</td>
</tr>
<tr>
<td>Poultry</td>
<td>1</td>
<td>-</td>
<td>0.7</td>
<td>-</td>
</tr>
<tr>
<td>Bread (cereal grains)</td>
<td>0.02</td>
<td>0.02</td>
<td>0.5</td>
<td>0.1</td>
</tr>
<tr>
<td>Potatoes</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
<td>-</td>
</tr>
<tr>
<td>Vegetables</td>
<td>-</td>
<td>0.1</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>Fruits</td>
<td>0.02</td>
<td>0.05</td>
<td>0.5</td>
<td>2</td>
</tr>
<tr>
<td>Sugar &amp; Adjuncts</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Oils</td>
<td>0.02</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Acceptable daily intake mg/kg body weight</td>
<td>0.002</td>
<td>0.0001</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>Acceptable daily intake for a person of 70 kg</td>
<td>0.014</td>
<td>0.007</td>
<td>0.7</td>
<td>1.4</td>
</tr>
</tbody>
</table>

* WHO value

REFERENCES


